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PATENT

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:  
KRULL et al.

Docket No.: 1997DE419/DIV

Serial No.: 09/956,544

Group Art Unit: 1713

Filed: September 18, 2001

Examiner: Zitomer, Fred

For: PROCESS FOR THE PREPARATION OF ETHYLENE COPOLYMERS, AND THEIR  
USE AS ADDITIVES TO MINERAL OIL AND MINERAL OIL DISTILLATESDECLARATION PER 37 C.F.R. §1.132Assistant Commissioner of Patents  
Washington, DC 20231

Dear Sir:

Applicants provide the following declaration evidence of Dr. Matthias Krull, co-inventor of the above-identified application for the purpose of traversing the rejection of claim 13 under 35 U.S.C. §103(a) as being unpatentable over Reimann et al., U.S. Patent No. 5,254,652 ("Reimann"), and also the rejection of claim 13 under 35 U.S.C. 103(a) as being unpatentable over Reimann taken with Ver Strate et al., U.S. Patent No. 4,804,794 ("Ver Strate").

This declaration provides objective evidence of unexpected results in the claimed invention to a process for improving cold flow of a mineral oil or a mineral oil distillate, set forth in detail in the claims and taught in the present specification.

I, Matthias Krull, am co-inventor of present Application Serial No. 09/956,544 as shown by the signed oath of record in this case. I have obtained the degree of Dr. rer. nat. (corresponds to Ph. D.) from the Free University Berlin, Germany, in 1989. I have been employed for 13 years in the Research and Development department of Hoechst AG, Frankfurt, Germany, which was succeeded by

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Clariant GmbH, Frankfurt, Germany, where my work has focused on oilfield chemicals and especially on cold flow additives for mineral oils.

I declare the following:

From my experience in the field of cold flow additives for mineral oils like fuel oil and diesel, a persistent problem occurs to researchers in the search for a new process for terpolymerization that gives terpolymers whose properties can be controlled better via the specific properties of the various monomers. However, it has surprisingly been found with the process of the present invention that improved polymers made from ethylene and at least two further comonomers can be obtained if the terpolymerization is carried out in a tubular reactor with one or more side branches and the fresh comonomers are fed in separately from one another via different reactor inlets.

The following experiments were conducted under my supervision to test the effectiveness of the copolymers investigated for improving the flow properties described with reference to the "cold filter point plugging test" (CFPP). The test was carried out in accordance with EN116. A detailed description of the essential methods followed in the experimental procedures is set forth below. A reactor as disclosed in Example 1 of the present application was used under the same reaction conditions as disclosed in Example 1. Five different terpolymers were prepared by changing the monomers, monomer concentration, and the feed procedure. Table 1 shows the conditions. The following abbreviations were used in the tables: VAc is vinyl acetate; MEK is methyl ethyl ketone; VeoVa® 10 (trademark of Shell Chemicals) stands for vinyl ester of versatic acid and is vinyl neodecanoate; and  $V_{140}$  is the viscosity measured at 140°C using a rotational viscometer in accordance with EN 3219.

Table 1

Example	Main feed	Side feed	Characterization of polymers
A	820 parts VAc	260 parts VeoVa® 10 130 parts MEK	29.5% VAc 9.2% VeoVa® 10 $V_{140} = 194 \text{ mPa}\cdot\text{s}$
B	650 parts VAc	280 parts 4-methyl-1-pentene 80 parts MEK	30.6% VAc 4.8% 4-methyl-1-pentene $V_{140} = 190 \text{ mPa}\cdot\text{s}$
Comparative C		260 parts VeoVa® 10 820 parts VAc 130 parts MEK	29.3% VAc 9.1% VeoVa® 10 $V_{140} = 205 \text{ mPa}\cdot\text{s}$
Comparative D		280 parts 4-methyl-1-pentene 650 parts VAc 80 parts MEK	30.4% VAc 4.8% 4-methyl-1-pentene $V_{140} = 204 \text{ mPa}\cdot\text{s}$
Comparative E	130 parts VeoVa® 10 410 parts VAc 65 parts MEK	130 parts VeoVa® 10 410 parts VAc 65 parts MEK	29.7% VAc 9.4% VeoVa® 10 $V_{140} = 190 \text{ mPa}\cdot\text{s}$

In Examples A and B, an ethylene/vinylacetate/ VeoVa® 10 terpolymer and an ethylene/vinyl acetate/4-methyl-1-pentene terpolymer were prepared according to the process of the present invention. In Comparative Examples C and D, identically constituted polymers (see feed composition and characterization of polymers in Table 1) were prepared by using only one feed which is in contrast to the process of the present invention. In Comparative Example E, a terpolymer was prepared according to a different process than the process of the present invention. The process differed in that both feeds contained all monomers in the same proportions. Comparative Example E was a comparative example for Example A. In Comparative Example E, half of the amounts of monomers that were fed into the single streams in Example A were fed into both streams.

The above-prepared terpolymers were used in a fuel having the properties set forth in

Table 2.

Table 2

initial boiling point	191°C
20% distillation	251°C
30% distillation	265°C
90% distillation	348°C
95% distillation	369°C
Cloud Point	+1°C
CFPP	-1°C
(90-20)%	97°C
S content	467 ppm

The filterability and cold flow properties of the fuel were determined to be as set forth in

Table 3.

Table 3

Terpolymer	ADT	CFPP with 100 ppm	CFPP with 200 ppm	CFPP with 300 ppm
Blank (no terpolymer added)	4.2	-1°C	-1°C	-1°C
Example A	6.8	-11°C	-15°C	-17°C
Example B	7.9	-9°C	-13°C	-16°C
Comparative Example C	45	-7°C	-12°C	-15°C
Comparative Example D	52	-8°C	-11°C	-14°C
Comparative Example E	7.2	-3°C	-4°C	-6°C

The ADT value was calculated in accordance with the equation set forth on page 16, lines 20-30 of the present specification. An ADT value of  $\leq 15$  is regarded as an indication that gas oil will have satisfactory use properties in "normal" cold weather. A product having an ADT value of  $> 25$  is referred to as non-filterable. As shown in Table 3, each of the ADT values obtained for Terpolymer C and Terpolymer D was considerably higher than 25 and, thus, was regarded as non-filterable. Terpolymer E, having an ADT value of  $\leq 15$ , did not render the oil non-filterable, but indicated that the CFPP performance was rather poor. Therefore, in view of the above and as one

of ordinary skill in the art, it is my opinion that the reactor type and the process of feeding different monomers through both main and side inlets are critical for the product of the polymerization process. Furthermore, it is also my opinion that these results obtained according to the process of the present invention were surprising.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Signed



Printed Name

DR. MATTHIAS KRAUS

Date

08.05.2003